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# Wind-hydrogen utilization for methanol production: An economy assessment in Iran

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ABSTRACT

This study investigates the feasibility to synthesis methanol from its flue gas and wind hydrogen. The concept is to mitigate $CO_2$ emission through flue gas recovery. Synthesizing methanol, on the other hand requires hydrogen at the rate of 3 kmol/kmol of carbon dioxide. Electrolysis is one method by which hydrogen can be produced cleanly from renewable source. Here it is assumed that the electrolysis unit is fed with the electricity from neighbor wind farms. Oxygen will be produced as a byproduct in electrolysis unit. However, electrolytic oxygen could be utilized for partial oxidation of methane in autothermal reactor (ATR). Onboard water electrolysis facilitates the oxygen and hydrogen storage, delivery and marketing. This study focuses on an integrated system of methanol production which enables green methanol synthesis through a system with zero carbon emission. Green methanol synthesis is comprised of $CO_2$ capturing and recycling along with renewable hydrogen generation. The produced hydrogen and $CO_2$ will be directed to methanol synthesis unit. By employing the integrated system for methanol synthesis, we could reduce the cost of using renewable energy technology.



**Contents**



## 1. Introduction

Converting carbon dioxide and hydrogen to methanol offers a great potential for cutting $CO_2$ emissions and the storage of hydrogen in a future energy scenario. Currently, most of the world's methanol is produced from natural gas via catalytic synthesis. Syngas-based production of methanol inevitably generates large amounts of carbon dioxide [1]. The concerns about rising fossil fuel prices, energy security and climate change have motivated the societies toward utilizing renewable energy sources, energy-efficient technologies, and carbon capture and storage method (CCS) to create a clean energy and sustained industry in the future.

South pars gas field, containing 7.5% of world's gas reservoirs and approximately half of Iran's gas reserves [2], is the economic hub of Iran. This region, however, is facing air pollution and environmental problems due to rapid growth of the gas and petrochemical industries. Hence, some measures should be taken to avoid excessive $CO_2$ emission into the atmosphere and remedy the environmental impacts in this area.

The objective of this study is to develop a supplementary system for chemical recycling of carbon dioxide to conventional methanol synthesis in Assalouyeh. This system consists of a flue gas recovery unit in which $CO_2$ will be separated from flue gas and an electrolysis unit with power input from the nearby wind farms. The produced

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carbon dioxide and hydrogen will be directed to methanol synthesis unit and will contribute in green methanol synthesis.

This scenario results in reducing the intensity of natural gas as the main feed of methanol plant; in addition, it mitigates $CO_2$ emission. By and large, an integrated system of methanol synthesis provides the opportunity for approaching zero-emission technology; without totally switching over renewable energy sources.

Iran has installed two mega-methanol plants in Assalouyeh under Lurgi license with overall nominal capacity of 10,000 ton/day. Utilizing the supplementary system of methanol synthesis appears to enhance sustainability of methanol technology and boost hydrogen technology in this region as well.

Methanol is a very powerful fuel that has an octane number of hundred. This means that methanol fuels allow for higher and more efficient compression than traditional gasoline. Methanol fuels including M85, burn much cleaner, with far fewer carbon emissions and particulates being released [3].

However, the magnitude of the project is constrained with the extent of available wind electricity. The feasibility of using wind power in Bardkhoun and Delvarfor for hydrogen generation in Assalouyeh was studied by Sayah A. which will be published later.

Chemical conversion of carbon dioxide to methanol has fascinated the world's attention toward improved methanol technology. Many studies have been made on this subject. Fujita S. et al explored methanol synthesis from $CO_2$ over Cu/ZnO catalysts; they have found that this catalyst exhibits high performance in synthesizing of methanol from $CO_2$ [4]. Mignard D. et al studied the feasibility of methanol synthesis from flue gas recovery and renewable electricity [5]. IAV and Carbon Recycling International have built a plant which uses electricity and $CO_2$ emissions from the geothermal power plant to produce renewable fuel for cars [6]. The Mitsui chemical has created catalyst adding variety of metals to copper and zinc base, and succeeded in synthesizing methanol efficiently from $CO_2$ and hydrogen. It has built a plant in Japan with a capacity of 100ton/year [7]. Scientists at the Singapore Institute of bioengineering and nanotechnology (IBN) have achieved an unparalleled feat by transforming carbon dioxide into methanol [8].

## 2. Hydrogen and methanol as alternative fuels

Because of the effect of carbon emissions into the atmosphere on global climate change, a carbon-constrained world is coming and alternative energy source will be required to supplement the carbon-intensive sources. One option to deal with the matter is the direct use of hydrogen [9]. Hydrogen conversion to heat and electricity is simple and clean. When hydrogen burns or combines with oxygen, it forms water and generates no pollutant. Nevertheless, hydrogen conversion to liquid methanol results in a more convenient alternative energy. Unlike hydrogen gas, liquid methanol is safe. Moreover it has the advantage of higher density for storage and transportation and it could also be handled by the existing infrastructure [10].

At any given pressure, hydrogen gas contains less energy per unit volume than liquid hydrocarbons like methane, methanol, ethanol, propane or octane (representing gasoline). Fig. 1 shows higher heating values (HHV) per unit volume of various energy carriers according to data extracted from "the engineering toolbox" [11]. Moreover, compared with other fuels, hydrogen combustion shows some disadvantages in that a large amount of its energy is inadvertently consumed to form water vapor what gets lost in flue gas. Although the analysis reveals that much more energy is needed to operate a hydrogen economy than is required for fossil energy supply and distribution today, precious amount of energy can be saved by packaging hydrogen chemically in a synthetic liquid hydrocarbon like methanol or ethanol. However, hydrogen

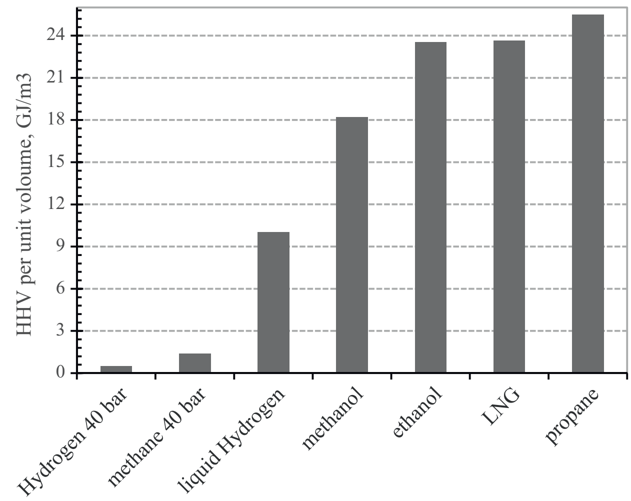



**Fig. 1.** Higher heating values (HHV) per unit volume of various energy carrier options.

production and transportation cost could be absorbed in the price of the synthesized chemicals [9,12–14].

Hydrogen can be produced from variety of sources including coal, natural gas, biomass, or water. Steam methane reforming (SMR) is currently the least expensive way to produce large quantities of hydrogen ($1.00–$5.00/kg of hydrogen). However a major drawback to using SMR is that the process is based on a nonrenewable fossil-fuel source [9].

However, to contribute in $CO_2$ mitigation, the scenario would require hydrogen from renewable source. This study focuses on wind hydrogen rather than other renewable energy sources with lower efficiency value. The renewable hydrogen will be used for methanol synthesis and will contribute in $CO_2$ recycling process. However, the resulting methanol could be regarded carbon neutral, as the hydrogen is produced cleanly and $CO_2$ is recycled [15]. Utilization of the improved methanol synthesis could moderate the cost of using clean hydrogen; hence, it appears to make $CO_2$ recycling economically more feasible.

Producing syngas from natural gas is a proven technology for methanol synthesis. Syngas-based methanol synthesis comprises three principles including: synthesis gas (syngas) production through steam reforming or partial oxidation of methane, catalytic conversion of the syngas to crude methanol according to equation (1) and (2) [16],and rectifying crude-methanol in distillation unit to obtain high grade methanol.

$$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O \tag{1}$$

$$CO + 2H_2 \rightarrow CH_3OH \tag{2}$$

To achieve the maximum yield in methanol synthesis, the stochiometery number (SN) presented in equation (3), shall be adjusted close to 2 [14,17,18].

$$SN = \frac{[H_2] - [CO]}{[CO] + [CO_2]} \tag{3}$$

To synthesize methanol from $CO_2$, $H_2/CO_2$ ratio should be adjusted close to 3. However, further studies may be required to improve catalyst selectivity toward $CO_2$ molecules.

## 3. Methodology

The scenario of methanol synthesis from its flue gas is presumed to comprise of three major principles including: carbon capturing, electrolytic hydrogen production from renewable energy sources,

**Table 1**
Methanol plant exhaust-gas specification.

| Specification | Unit | Stream 1 | Stream 2 |
|---|---|---|---|
| Flow | Sm3/h | 152,458 | 171,785 |
| Temperature | °C | 141 | 181 |
| Pressure | bar | 1 | 1 |
| $CO_2$ | Mol% | 10.74 | 7.92 |
| $N_2$ | Mol% | 65.73 | 65.87 |
| $O_2$ | Mol% | 1.53 | 2.09 |
| $H_2O$ | Mol% | 21.18 | 23.3 |

and finally catalytic synthesis of methanol from $CO_2$ and hydrogen in a conventional methanol synthesis unit.

### 3.1. $CO_2$ capturing

Amine absorption is a proven technology for carbon dioxide recovery from flue gases. Several researchers have modeled and studied $CO_2$ capturing by mono-ethanol amine (MEA). Rochelle G.T. found 5-20% reduction in energy requirement where a multi-pressure stripping was employed. He considered achieving temperature approach of 50C within the heat exchanger [19].

Singh, D. et al found that the thermal energy requirement for $CO_2$ capturing in a coal fired power of 400 MW is equal to 3.8 GJ/ton $CO_2$ [20]. Alie, C. et al investigated the lowest energy requirement of 176 kJ/kmol $CO_2$ (4GJ/ton $CO_2$) can be achieved at lean solvent loading between .25-.3 mol $CO_2$/mol MEA [21]. Abu-Zahra M. et al realized the energy amount of 3.3 GJ/ton $CO_2$ is required for solvent regeneration with 30 wt% MEA [22]. The phenomenon of $CO_2$ absorption in MEA can be described by electrochemical reaction in the aqueous solution according to Eqs (4–7) [22].

$$CO_2 + 2H_2O \Leftrightarrow H_3O^+ + HCO_3^- \quad (4)$$

$$HCO_3^- + H_2O \Leftrightarrow H_3O^+ + CO_3^{2-} \quad (5)$$

$$MEA + H_3O^+ \Leftrightarrow MEA^+ + H_2O \quad (6)$$

$$MEA + HCO_3^- \Leftrightarrow MEACOO^- + H_2O \quad (7)$$

The procedure is like that first, the flue gas gets cooled and dehumidified. By passing through a separator, it becomes enriched of $CO_2$. Afterwards, the $CO_2$-rich flue gas flows through an absorption column counter currently with MEA. $CO_2$ gas gets absorbed in MEA and leaves the column via its bottom, while the top product ($N_2$) is vented to atmosphere. Finally, MEA and $CO_2$ get separated by distillation. Here we consider regeneration energy demand equal to 3.5 GJ/ton $CO_2$ due to higher MEA temperature.

Utilizing thermal energy of flue gas could reduce the cost for $CO_2$ capturing process.

Table 1 shows specification of studied exhaust gas.

If $CO_2$ capturing with the recovery of 96% is planned, 29.26 ton/h and 24.30 ton/h $CO_2$ will be captured from stream 1 and stream 2 respectively. To provide a quantitative basis for reducing the emissions, Mckinsey has developed global greenhouse gas abatement data base which summarizes the technical opportunities to reduce emissions at a cost of up to 60 \$/t$CO_2$ of avoided emissions.

To evaluate the economy for the topic we consider its data for CCS of 20–30 €/ton $CO_2$ [23].

### 3.2. Hydrolysis

Hydrogen can be produced cleanly via water electrolysis powered by electricity from renewable source like solar radiation, wind kinetic energy, water, etc [24,25]. It is used for production of the liquid fuels like methanol and di-methyl ether. Hence, hydrogen is an energy carrier rather than a primary energy source. It counts as main link between intermittent renewable sources and chemical energy [10].

Electrolysis uses direct current (DC) electricity to split water into its basic elements of hydrogen and oxygen. Since the process uses only water as a source, it can produce up to 99.9995% pure hydrogen and oxygen. Electrolysis was discovered in 1800 by William Nicholson and Sir Anthony Carlisle shortly after Alessandro Volta invented the electric battery.

Several electrolytic processes are developed including: alkaline and polymer electrolyte membrane (PEM), high temperature decomposition, photo-electrolysis (photolysis), photo-biological production (bio-photolysis). However, amongst them, alkaline electrolysis and PEM are commercially available. PEM electrolysis is suitable for small capacities while alkaline have dominated high-capacity industrial market [9]. Indeed alkaline electrolysis was adopted for hydrogen production in this study. Alkaline electrolysis uses an aqueous KOH solution as an electrolyte that usually circulates through the electrolytic cells [9,25]. The principle of alkaline electrolysis is presented in Eqs. (8–11) [26].

$$\text{Electrolyte}: \quad 4H_2O \rightarrow 4H^+ + 4OH^- \quad (8)$$

$$\text{Cathode}: \quad 4H^+ + 4e^- \rightarrow 2H_2 \quad (9)$$

$$\text{Anode}: \quad 4OH^- \rightarrow O_2 + 2H_2O + 4e^- \quad (10)$$

$$\text{Sum}: \quad 2H_2O \rightarrow O_2 + 2H_2 \quad (11)$$

The produced $H_2$ and $O_2$ will be transferred into storage tanks to be used later. $O_2$, however, is then routed to autothermal reactor (ATR) and contributes to the partial oxidation of methane (POX). Onsite consumption of oxygen lessens the difficulty of its market and dispatch.

The equation for the water electrolysis reaction is the reverse of the fuel cell equation.

$$H_2O + 237.2\,\text{kJ/mole electricity} + 48.6\,\text{kJ/mole heat} \rightarrow H_2 + \tfrac{1}{2}O_2 \quad (12)$$

The efficiency of an electrolysis system may be expressed as the heating value of hydrogen produced divided by the electrical energy input.

$$\text{Electricity efficiency}_{(HHV)} = \frac{\text{HHV of hydrogen produced}}{\text{Electricity used}} \quad (13)$$

However, since both electricity and heat are used to split water electrochemically using this approach to calculate the maximum thermodynamic efficiency produces nonsense numbers that exceed 100%. Electrolysis efficiency can be assessed through voltage efficiency of cells which is defined as the actual cell operating voltage divided by the thermodynamic cell voltage.

$$\text{Voltage efficiency} = \frac{\text{Operating voltage} \quad (V)}{\text{Thermodynamic voltage} \quad (E)} \quad (14)$$

Total energy of reaction is composed of both electrical and thermal energy known as enthalpy ($H$). The amount of electrical energy is known as the Gibbs free energy ($G$) and corresponds to maximum amount of useable electrical energy available when hydrogen recombines with oxygen. Heat of reaction can be translated into irreversible energy or entropy ($S$). The thermodynamic cell voltage can be calculated using the Gibbs free energy and the Nernst equations.

Gibbs free energy can be converted into a thermodynamic voltage using the formula in which free energy in joules per mole, as shown below:

$$G = nFE^0 \quad (15)$$

Under standard conditions, the Gibbs free energy for the hydrogen–oxygen reaction is 237.2 kJ/mole for production of the

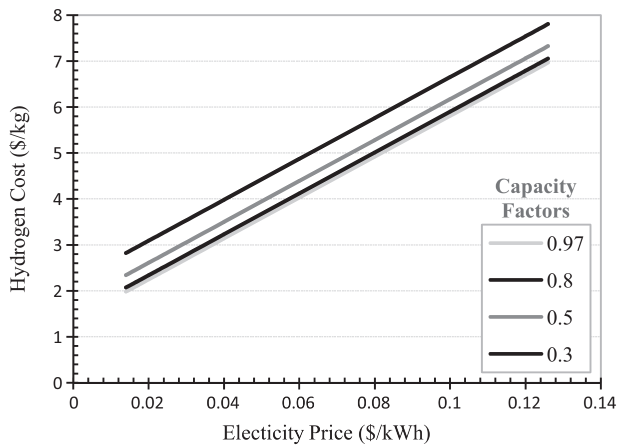



**Fig. 2.** Future wind hydrogen production system: capacity factor and electricity price effect on hydrogen cost in $/kg (76% LHV efficiency, 10% IRR for a 180 $/kW electrolyzer installed and 1.00 $/GGE hydrogen delivery cost).

liquid water at 25 °C. Therefore, the thermodynamic voltage for a hydrogen–oxygen fuel cell operating at standard temperature and pressure is 1.229 V.

$$E^0 = \frac{237,200\,\text{J}}{2(96,485)} = 1.229\,\text{V}$$

The approximate efficiency for an electrolysis cell stack working under standard conditions can be calculated by dividing the operating voltage by the standard thermodynamic voltage $E^0$ [27].

Splitting a mole of liquid water to produce a mole of hydrogen at 25 °C requires 285.8 kJ of energy. Letting this value in Eq. (15) results in a cell voltage of 1.481 V. It is the voltage at which an electrolysis cell, running at 25 °C, can operate without producing excess heat.

$$\text{Voltage efficiency} = \frac{1229}{1481} = .83$$

Therefore; there is a maximum theoretical limit to the electrical efficiency attainable by a fuel cell system.

## 4. Wind-hydrogen capacity and economy

Ideally, 285.8 kJ energy per mol of hydrogen, 39 kWh/kg of hydrogen, is required to split water. Commercial electrolyzer units have lower efficiency and require 54–67 kWh/kg of hydrogen produced [9]. The expenditure for hydrogen production consist of capital investment, electricity cost and operating & manual (O&M) cost. The price of electricity and capital cost are the most significant factors in cost of hydrogen via electrolysis [25].

At large and small installation size, the significance of capital cost and electricity cost on hydrogen cost are reversed. For the larger sizes (1000 kg/day) electricity is the biggest factor, amounting to more than 62% of the total cost, and capital cost could be closer to 30%. This trend is nearly reversed at the smaller sizes (10 kg/day), for which capital cost has biggest effect at 62% of final cost, with electricity cost at 20% [25]. However, previous studies show that there is a potential market for wind/hydrogen energy systems in remote areas and weak grids [28,29].

The U.S. department of energy (DOE) aims to achieve higher system efficiencies in the future (by year 2030) with 44.5 kWh/kg electricity consumption or 76% LHV efficiency. It has set the cost target for hydrogen at $2.0-3.0/kg to serve the transportation market [24,25,27]. Fig. 2 shows how delivered hydrogen price changes at different capacity factors and electricity prices for a future case where electrolyzer cost and delivery costs have decreased and electrolyzer efficiencies has increased [8].

**Table 2**
Utilities and future expenditure for methanol production from flue gas and wind hydrogen.

| Utility | Unit cost | Total cost ($/ton Meoh) |
|---|---|---|
| Hydrogen | 1 $/kg = 2 $/kmol | 187.5 |
| $CO_2$ capturing | 30 $/ton | 41.25 |

The higher capacity factor cases here allow for wind farms that have the possibility of buying electricity from the grid so that electrolyzer can operate at its maximum operating capacity factor of 97%. Higher electricity prices represent either higher wind costs, higher grid electricity costs, or a combination of two. The higher capital cost is the more O&M would be. According to DOE statements, hydrogen produced from wind has the potential to meet cost goals of $2.00–3.00/kg, including production, delivery, and dispensing [9].

Considering centralized hydrogen production and use at methanol plant would eliminate delivery cost and hence results in lower hydrogen price to $1–2/kg.

If employment of an improved electrolysis system with power consumption of 44.5 kWh/kg of hydrogen is assumed; then, the scenario would require power supply of 177.6 MWh and 147.5 MWh to generate enough hydrogen for chemical recycling of $CO_2$ from stream 1 and stream 2.

## 5. Results and discussion

Table 2 shows the unit cost for utilities involved in methanol synthesis and the future utility expenditure per unit methanol produced from fuel gas and wind hydrogen.

In this scenario we regard methane price equal to 4.48 $/kg by 2030. It means that the hydrocarbon cost for conventional methanol synthesis will be 133.47 $/ton methanol. Running a stand alone green methanol synthesis from flue gas and wind hydrogen leads on raising material cost by a factor of 1.71% compared with methanol synthesis from natural gas. However, from flue gas and wind hydrogen using the integrate system moderate it to a material cost of 1.11%. By operating the integrated system, about 0.19 ton of green methanol could be produced per ton fossil based methanol, that means lowering methane intensity as feed gas to methanol plant. Running the integrated system makes around .42 Mton $CO_2$ emission avoided per year.

It appears that the scenario is compatible with the conventional methanol system. Furthermore, taking the real impact of fossil fuels on the environment to consideration, if $CO_2$ abatement cost will be raised, it would make the process even economically more viable.

## 6. Conclusion

0.27 ton carbon dioxide is emitted per ton methanol synthesized in the studied methanol plant. Utilizing this amount of $CO_2$ for green methanol synthesis in an integrated system reduces methane intensity at a rate of 0.11 ton per ton of methanol produced, provided that $CO_2$ selective catalysts are supplied and well configured inside methanol synthesis reactors.

Although great concern is growing for emission of greenhouse gases, switch to renewable energy source is not possible in short term. While developing an integrated system seems to be a reasonable solution to the problem.

The studied scenario would require 325.1 MWh power energy at a cost of .01 $/kWh to be used in electrolysis system with energy use of 44.5 kWh/kg of $H_2$ generated. However, proportionate to the availability of electricity, the project could be scaled to a lower capacity.

On the other hand, further studies shall be carried out on methanol synthesis catalyst which could enhance carbon dioxide reactivity with hydrogen.

All in all, running the integrated system of methanol synthesis has the potential to cut $CO_2$ emissions around 0.42 Mton annually. The average material cost for methanol depends on whether a separate unit for carbon dioxide to methanol synthesis is applied or not. Regardless of other energy saving potential like lowering steam and methane intensity; we estimate that a combination of green and fossil based methanol synthesis results in adding to material cost not more than 11% compared to conventional methanol synthesized unit. Hence, if renewable electricity and electrolyzer with higher efficiency would be available so that hydrogen could be produced at $1-2/kg, the improved system of methanol synthesis appears to be viable.

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